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## **JAPANESE / ENGLISH TRANSLATION OF**

**Japanese Patent Application JP 2000 - 186191 A**

**Noncrystalline Polyethylene Terephthalate Resin Composition for  
Calendering**

**Your Ref: 1022-03-21**

**For: Eastman Chemical Company**

9109 Eagle Hills Dr . Las Vegas NV 89134 : ph: 866-722-7990 (toll-free) . fax: 702-973-6009 . email: sales@tls-translations.com

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(21) Application No.: 10-363908	(71) Applicant: 000190116 Shin-Etsu Polymer Co., Ltd. 3-5 Nihonbashi 4-chome, Chuo-ku, Tokyo- to
(22) Date of Filing: December 22, 1998	(71) Applicant: 000186810 Akishima Chemical Industries, Co., Ltd. Bungei Shunju Shinkan 3-23 Kioi-cho, Chiyoda-ku, Tokyo-to
	(72) Inventor: Nobumitsu Suda c/o Shin-Etsu Polymer Co., Ltd., Tokyo Plant, 406-1 Yoshino-cho 1-chome, Omiya-shi, Saitama- ken
	(74) Agent: 100062823 Ryoichi Yamamoto, Patent Attorney (and 2 others)
(Continued on last page)	

**(54) [Title of the Invention]      Noncrystalline Polyethylene Terephthalate Resin  
Composition for Calendering**

**(57) [Summary]**

**[Object]** To provide a noncrystalline polyethylene terephthalate sheet resin composition that can be easily released by metal rolls adjusted to a high temperature during calendering, has superior transparency and surface smoothness, is free from flow marks and the like, and can yield molded sheets with extremely high productivity.

**[Means of Achievement]** The aforementioned resin composition is obtained by means of compounding of 0.1 to 5 parts by weight of either an organic phosphoric acid ester or a fatty acid ester, or both, per 100 parts by weight of noncrystalline polyethylene terephthalate resin.

**[Claims]**

**[Claim 1]** A noncrystalline polyethylene terephthalate resin composition for calendering, characterized in that 0.1 to 5 parts by weight of either a phosphoric acid ester or a fatty acid ester, or both, is compounded per 100 parts by weight of a noncrystalline polyethylene terephthalate resin.

**[Claim 2]** A noncrystalline polyethylene terephthalate resin composition for calendering as set forth in Claim 1, wherein the organic phosphoric acid ester is a compound represented by the general formula  $R^1O(CH_2CH_2O)_n PO[O(CH_2CH_2O)_m R^2]_{2-Y} (OH)_Y \cdots$  (wherein,  $R^1$  and  $R^2$ , which may be the same or different, indicate an alkyl or alkali group with 4 to 24 carbon atoms; Y indicates an integer of 1 or 2; and n and m indicate, respectively, integers of 1 to 100).

**[Claim 3]** A noncrystalline polyethylene terephthalate resin composition for calendering as set forth in Claim 2, wherein the organic phosphoric acid ester is a nonyl phenyl polyoxyethylene phosphoric acid ester having different polyoxyethylene groups in which n and m in the aforementioned general formula are 5 to 55, respectively, or a tridecyl polyoxyethylene phosphoric acid ester having different polyoxyethylene groups in which these are 4 to 10, respectively.

**[Claim 4]** A noncrystalline polyethylene terephthalate resin composition for calendering as set forth in Claim 1, wherein the fatty acid ester is a synthetic wax or natural wax comprised of an aliphatic saturated carboxylic acid with 12 to 28 carbon atoms and an aliphatic saturated alcohol with 2 to 30 carbon atoms.

**[Claim 5]** A noncrystalline polyethylene terephthalate resin composition for calendering as set forth in Claim 4, wherein the fatty acid ester is a montanic acid glycol ester, a montanic acid glyceride, or montan wax.

**[Detailed Description of the Invention]**

**[0001]**

**[Technological Field of the Invention]** The present invention relates to a noncrystalline polyethylene terephthalate resin composition of superior calender processability.

**[0002]**

**[Prior Art]** Polyethylene terephthalate sheets are generally used as packaging materials for food products and as laminated sheets for construction, household electrical implements, and automotive applications. The polyethylene terephthalate sheets that are used in these fields are manufactured conventionally by means of extrusion molding and injection molding. In these methods, a die is adjusted to a specified thickness, after which withdrawal processing is performed as the molten resin is discharged and rapidly cooled from the softening temperature of the resin. While release from the withdrawal roller or the mold can be performed easily, components in which there are localized, marked differences in thickness often occur even if the die is adjusted. Such sheets are unsuitable for printing and for processing as laminates and coatings. This is a cause of development of holes when the sheets are subjected to secondary molding as blister packs, trays, or carrier tape.

**[0003]** In comparison with these molding methods, calendering is a method that is superior for mass production and allows molten resin to be made into sheets of the desired thickness by means of rolling with a heated metal roller. Superior product quality is therefore obtained in the sense that no difficulties are encountered in the vicinity of the die, there is little occurrence of localized differences in thickness, and the method is suited to manufacturing products of similar specifications in large volume and has come to be widely used for molding and processing sheets made of commonly used resins such as polyvinyl chloride.

**[0004]** However, even when noncrystalline polyethylene terephthalate resins are used in the same calendering method, there is poor release from the metal roller and processing is difficult. For this reason, improvements have been attempted by means of adding various types of additives to the resin. However, sufficient release effectiveness cannot be obtained with small quantities. When large quantities are added, there is the problem that the resulting sheet may have poor transparency. Thus, noncrystalline polyethylene terephthalate resins for calendering that meet the requirements of both release properties and transparency have not yet been obtained.

**[0005]**

**[Problems to Be Solved by the Invention]** Consequently, the object of this invention is to obtain a noncrystalline polyethylene terephthalate resin composition for calendering (hereafter referred to simply as noncrystalline polyethylene terephthalate resin composition) that can be

easily released by metal rolls adjusted to a high temperature during calendering, has superior transparency and surface smoothness, is free from flow marks and the like, and can yield molded sheets with extremely high productivity.

[0006]

**[Means Used to Solve the Above-Mentioned Problems]** The noncrystalline polyethylene terephthalate resin composition of this invention is a substance obtained by means of compounding 0.1 to 5 parts by weight of either a phosphoric acid ester or a fatty acid ester, or both, per 100 parts by weight of a noncrystalline polyethylene terephthalate resin, and, preferably, it is a substance in which the organic phosphoric acid ester is a compound represented by the general formula  $R^1O(CH_2CH_2O)_n PO[O(CH_2CH_2O)_m R^2]_{2-Y} (OH)_Y \dots$  (wherein  $R^1$  and  $R^2$ , which may be the same or different, indicate an alkyl or alkali group with 4 to 24 carbon atoms; Y indicates an integer of 1 or 2; and n and m indicate, respectively, integers of 1 to 100), and in particular, a nonyl phenyl polyoxyethylene phosphoric acid ester having different polyoxyethylene groups in which n and m in the aforementioned general formula are 5 to 55, respectively, or a tridecyl polyoxyethylene phosphoric acid ester having different polyoxyethylene groups in which these are 4 to 10, respectively, and in which the fatty acid ester is a synthetic wax or natural wax comprised of an aliphatic saturated carboxylic acid with 12 to 28 carbon atoms and an aliphatic saturated alcohol with 2 to 30 carbon atoms, and, in particular, is a montanic acid glycol ester, a montanic acid glyceride, or montan wax.

[0007]

**[Embodiment of the Invention]** The noncrystalline polyethylene terephthalate resin of this invention should be a transparent substance of which the principal components may, for example, be 100 mol % of terephthalic acid, 30 to 90 mol % of ethylene glycol, and 10 to 70 mol % of 1,4-cyclohexane dimethanol, with substances comprised of 100 mol % of terephthalic acid, 65 to 80 mol % of ethylene glycol, and 20 to 35 mol % of 1,4-cyclohexane dimethanol being particularly desirable from the standpoint of calender processability.

[0008] The organic phosphoric acid ester is a compound represented by the general formula  $R^1O(CH_2CH_2O)_n PO[O(CH_2CH_2O)_m R^2]_{2-Y} (OH)_Y \dots$  (wherein  $R^1$  and  $R^2$ , which may be the same or different, indicate an alkyl or alkali group with 4 to 24 carbon atoms; Y indicates an integer of 1 or 2; and n and m indicate, respectively, integers of 1 to 100). They may, for example, be octyl

polyoxyethylene (n, m = 5 to 55, indicating mixed esters having different polyoxyethylene groups in which n and m are 5 to 55; the same hereafter) phosphoric acid esters, decyl polyoxyethylene (n, m = 5 to 25) phosphoric acid esters, dodecyl polyoxyethylene (n, m = 4 to 10) phosphoric acid esters, tridecyl polyoxyethylene (n, m = 4 to 10) phosphoric acid esters, octyl phenyl polyoxyethylene (n, m = 5 to 55), nonyl phenyl polyoxyethylene (n, m = 5 to 55) phosphoric acid esters, and dodecyl phenyl polyoxyethylene (n, m = 5 to 55) phosphoric acid esters. Of these, dodecyl polyoxyethylene (n, m = 4 to 10) phosphoric acid esters and nonyl phenyl polyoxyethylene (n, m = 5 to 55) phosphoric acid esters are preferable.

[0009] The fatty acid esters are synthetic waxes or natural waxes comprised of aliphatic saturated carboxylic acids with 12 to 28 carbon atoms, and aliphatic saturated alcohols with 2 to 30 carbon atoms. The aliphatic saturated carboxylic acids with 12 to 28 carbon atoms that constitute the aforementioned synthetic waxes include, for example, lauric acid, myristic acid, stearic acid, behenic acid, lignoceric acid, cerotic acid, and montanic acid, whereas the aliphatic saturated alcohols with 2 to 30 carbon atoms include, for example, ethyl, octyl, lauryl, myristyl, stearyl, behenyl, pentacosyl, ceryl, octacosyl, and melissyl alcohols as well as polyvalent alcohols such as polyethylene glycol and glycerol.

[0010] The aforementioned waxes include, for example, stearyl laurate, stearyl myristate, stearyl stearate, octyl behenate, lauryl behenate, myristyl behenate, stearyl behenate, behenyl behenate, pentacosyl behenate, ceryl lignocerate, octacosyl lignocerate, myristyl lignocerate, stearyl cerotate, behenyl cerotate, ceryl cerotate, melissyl cerotate, ethyl montanate, ceryl montanates, and montanic acid glycol esters. The natural waxes include montan wax, carnauba wax, beeswax, candelilla wax, rice bran wax, and insect wax. Of these, montanic acid glycerol esters, montanic acid glycerides, and montan wax are preferable.

[0011] The noncrystalline polyethylene terephthalate resin composition of this invention is compounded by means of adding 0.1 to 5 parts by weight, and, preferably, 0.2 to 3 parts by weight, of either an organic phosphoric acid ester or a fatty acid ester, or both, per 100 parts by weight of noncrystalline polyethylene terephthalate resin composition. When the quantity added is less than 0.1 parts by weight, there is no effect on the release from the metal roller for calendering, and there is poor bank rotation. When more than 5 parts by weight are added, sheet transparency is lost and there is also loss of surface smoothness, which is undesirable.

[0012] In addition, stabilizers, surfactants, antioxidants, antistatic agents, fillers, ultraviolet absorbers, and colorants can be added as required to the aforementioned noncrystalline polyethylene terephthalate resin composition in addition to the aforementioned components in a range that does not impair calendering capacity and transparency.

[0013] The noncrystalline polyethylene terephthalate resin composition of this invention and the aforementioned components are mixed with a mixer, after which the mixture is kneaded with an extruding machine and may be made into a sheet using a calendering machine. At this time, the aforementioned sheet-shaped material can be released from the roller extremely easily; flow marks, skin roughness, and air marks are not produced; and a sheet-shaped material of superior external appearance can be processed with high productivity.

[0014]

[Working Examples] We shall now describe specific embodiments of this invention by presenting working examples and comparative examples.

**(Working Examples 1 to 5 and Comparative Examples 1 to 6)** The materials indicated below were added in the amounts (parts by weight) shown in Table 1 to 100 parts by weight of noncrystalline polyethylene terephthalate resin, and the materials were mixed to a homogeneous state with a Henschel mixer. Kneading was performed by means of an extruding machine up to a resin temperature of 200°C, with the noncrystalline polyethylene terephthalate resin composition of this invention being prepared. A backward L-shaped four-roller calendering machine in which the surface temperatures of the first roller and second roller were set to 210°C, the surface temperature of the third roller was set to 200°C, and the surface temperature of the final roller was set to 195°C was used to process a sheet-shaped material of 0.3 mm in thickness and 1,000 mm in width. At this time, the haze (haze value), release from the metal roller, and the surface smoothness of the resulting sheet were evaluated on the basis of the standards indicated below, and the results are shown in Table 1.

[0015] (Materials Used)

Noncrystalline polyethylene terephthalate resin: PETG copolyester 6763 (product name, manufactured by the Eastman Chemical Company)

Organic phosphoric acid ester A: No. 1737 [product name; tridecyl polyoxyethylene (n, m = 4 to 10) phosphoric acid ester, manufactured by Akishima Chemical Industries]

Organic phosphoric acid ester B: nonyl phenyl polyoxyethylene (n, m = 5 to 55) phosphoric acid ester

Fatty acid ester A: Hostalub WE4 (brand name, montanic acid ethylene glycol ester, manufactured by Clariant Japan)

Fatty acid ester B: Montanic acid glyceride

Acrylic acid resin: L-1000 (brand name, manufactured by Mitsubishi Rayon Co., Ltd.)

[0016] (Evaluation Standards)

“Haze (haze value)”

Haze (haze value) of a sheet with 0.3 mm in thickness determined in accordance with JIS K 7105

40 or less	⊙
40 to 60	O
60 or higher	X

[0017] “Release from metal roller”

Good release of molten and softened sheet from metal roller of calender O

Poor release of molten and softened sheet from metal roller of calender X

[0018] “Smoothness of sheet surface”

Surface of sheet was smooth and level O

Surface of sheet was rough and not level X

[0019]

Table 1

		Working Examples					Comparative Examples					
		1	2	3	4	5	1	2	3	4	5	6
Noncrystalline polyethylene (Pbw) Terephthalate resin		100	100	100	100	100	100	100	100	100	100	100
Organic phosphoric acid (Pbw) Ester A		1	0	0	0	0.5	0	0.08	0	7	0	0
Organic phosphoric acid (Pbw) Ester B		0	1	0	0	0	0	0	0	0	7	0
Fatty acid ester A (Pbw)		0	0	1	0	0.5	0	0	0.08	0	0	0
Fatty acid ester B (Pbw)		0	0	0	1	0	0	0	0	0	0	0
Acrylic acid resin (Pbw)		0	0	0	0	0	0	0	0	0	0	3
Results of eval- uation	Haze (haze value)	⊙	○	⊙	○	⊙	⊙	⊙	⊙	X	X	X
	Release from metal roller	○	○	○	○	○	X	X	X	○	○	○
	Smoothness of sheet surface	○	○	○	○	○	X	X	X	X	X	○

Pbw = parts by weight

[0020]

**[Effect of the Invention]** The noncrystalline polyethylene terephthalate resin composition of this invention can be easily released by metal rolls adjusted to a high temperature during calendering, has superior transparency and surface smoothness, is free from flow marks and the like, and can yield molded sheets with extremely high productivity.

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(72) Inventor: Yusuke Tsukagoe  
c/o Shin-Etsu Polymer Co., Ltd., Tokyo Plant  
406-1 Yoshino-cho 1-chome, Oshima-shi, Saitama-ken

(72) Inventor: Yasumoto Kodana  
c/o Shin-Etsu Polymer Co., Ltd., Tokyo Plant  
406-1 Yoshino-cho 1-chome, Oshima-shi, Saitama-ken

(72) Inventor: Hideichi Tsukida

Room 202, Fussa Cosmo Heights, 231-1 Shimo, Fussa-shi,  
Tokyo-to

(72) Inventor: Gimer Fabrice

Room 302, SR Building, 22-18 Asahi-cho 4-chome,  
Akishima-shi, Tokyo-to

(72) Inventor: Masaaki Itakura

126-61 Wagahara 1-chome, Tokorozawa-shi, Saitama-ken

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